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High-Field Magnetoresistance of $La_{0.67}Sr_{0.33}MnO_3$ Thin Films Deposited on $LiNbO_3$ Substrates

I.T. Gomes · A.M. Pereira · J. Ventura · F. Carpinteiro · J.M. Moreira · J.P. Araújo · J. Barbosa · B.G. Almeida

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Abstract Colossal magnetoresistive manganites have been widely studied due to their potential use in sensor and device applications. In this work, $La_{0.67}Sr_{0.33}MnO_3$ thin films were deposited by pulsed laser ablation on $LiNbO_3$ substrates and magnetoresistance measurements were performed using pulsed magnetic fields up to 25 T. The corresponding magnetoconductance of the films was fitted in order to obtain the grain boundary (GB) contribution to the transport properties. The observed temperature dependence of the fitting parameters was indicative of antiferromagnetism across GB spins and reflected the progressive reduction of magnetic ordering with increasing temperature.

Keywords Lanthanum strontium manganite · Pulsed laser ablation · Magnetoresistance · High magnetic fields

1 Introduction

Rare-earth manganites exhibiting colossal magnetoresistance (CMR) have been attracting much scientific and technological interest [1, 2]. In the perovskite-type system $La_{1-x}Sr_xMnO_3$ (LSMO), a ferromagnetic coupling between the Mn ions develops in the concentration range $x = 0.2$–0.6, due to the double exchange mechanism [2]. Near the Curie temperature (T_C) the electrical resistivity (ρ) has a maximum

I.T. Gomes · J. Barbosa · B.G. Almeida (✉)
Departamento de Física, Universidade do Minho, Campus de Gualtar, 4710-057 Braga, Portugal
e-mail: bernardo@fisica.uminho.pt

A.M. Pereira · J. Ventura · F. Carpinteiro · J.M. Moreira · J.P. Araújo
Departamento de Física and IFIMUP-IN, Institute of Nanoscience and Nanotechnology, Universidade do Porto, Rua do Campo Alegre 687, 4169-007 Porto, Portugal

and, below a characteristic metal-insulator transition temperature (T_{MI}), a metallic-like ρ-behavior is observed. On LSMO, T_C can attain relatively high values by changing the Sr concentration, reaching ∼380 K for $x = 1/3$ [2]. Combined with the CMR property, such high T_C makes LSMO a promising material for room-temperature magnetic sensors, recording devices or electrodes in ferroelectric memories [1, 2]. It is known that grain boundary scattering plays an important role on the electrical properties of LSMO samples [1]. The choice of substrate is also determinant in the obtained transport and magnetic properties of LSMO thin films [2]. In this regard, lithium niobate ($LiNbO_3$) is widely used in electro-optical devices [3]. Thus, in this work, it is shown, for the first time, that highly oriented LSMO thin films can be deposited by pulsed laser ablation on $LiNbO_3$ crystals. Their transport properties were characterized in the 77–300 K temperature range and in magnetic fields up to 25 T.

2 Experimental Details

The $La_{0.67}Sr_{0.33}MnO_3$ thin films were prepared by pulsed laser ablation on z-cut $LiNbO_3$ crystal substrates. The depositions were done with a KrF excimer laser (wavelength $\lambda = 248$ nm), at a fluence of 2 J/cm^2, with a 3 Hz repetition rate and pulse duration of 25 ns. The oxygen pressure during preparation was 0.5 mbar and the substrate temperature was 720°C. After the depositions, the films were cooled down to room temperature at atmospheric oxygen pressure.

The magnetoresistance (MR) was measured in a pulsed magnetic field unit providing magnetic fields up to 25 T (rising time 0.1 s; discharge time 2 s) and an energy of 0.6 MJ/pulse. The MR-setup uses a low distortion a.c. generator (0–200 kHz frequency range, 20 μVpp–40 Vpp output voltage), and a high resolution lock-in amplifier. Both the real and imaginary signals are then acquired with a fast 16-bit board (333000 points per second). The magnetoresistance of the LSMO film was measured between 77 K and 300 K, with the magnetic field parallel to the electrical current, using a standard four probe method.

3 Results and Discussion

X-ray diffraction measurements performed on the thin film samples (with a Siemens D5000 diffractometer using Cu K_α radiation) revealed that the films are (111) oriented and present grain sizes ∼235 nm (not shown). Their pseudocubic lattice parameter along the growth direction was $a_{film} = 3.86$ Å, which was lower than the bulk ($a_{bulk} = 3.873$ Å), and the samples thickness, estimated by scanning electron microscopy, was approximately 400 nm. The Curie temperature measured with a Superconducting Quantum Interference Device (SQUID) magnetometer was $T_C \sim 360$ K.

Figure 1 shows the temperature dependence of the electrical resistivity [ρ(T)], measured in the 10–300 K range. The resistivity initially rises, attaining a maximum at the metal-insulator transition temperature $T_{MI} \sim 260$ K. Then, below T_{MI}, ρ changes to a metallic-like behavior characteristic of the ferromagnetic phase. The presence of spin-dependent tunneling of electrons across grain boundaries (GB) on

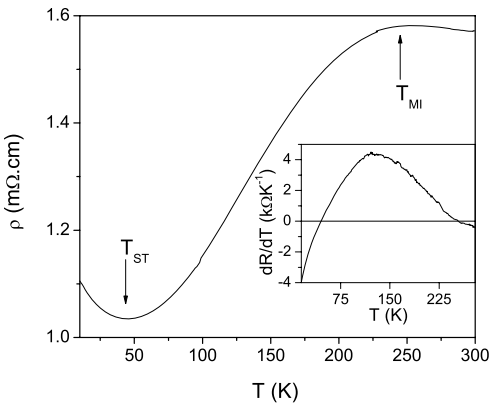

Fig. 1 Temperature dependence of the electrical resistivity measured on a laser ablation deposited LSMO thin film. *Inset*: Temperature derivative dR/dT

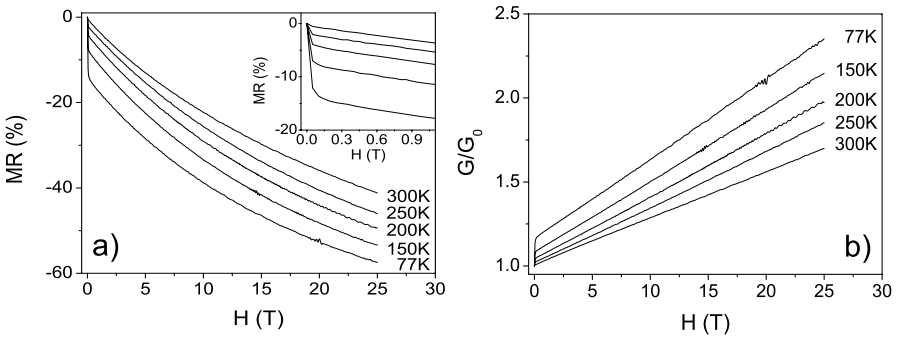

Fig. 2 (**a**) Magnetoresistance and (**b**) magnetoconductance of a laser ablation deposited LSMO thin film. Inset displays an enlarged view of the low field region

the films [1, 2] again leads to a resistivity increase below $T_{ST} \sim 50$ K. This is in agreement with the observed lower T_C as compared to the bulk ($T_{C,bulk} = 380$ K).

Figure 2(a) shows the magnetoresistance (MR) of the LSMO films, measured in the 77–300 K temperature range. For low fields [$H \leq 0.1$ T, inset of Fig. 2(a)] one observes an initial sharp decrease of the MR, due to spin-dependent tunneling across GB [2]. For higher fields, a more gradual MR decrease is observed. Figure 2(b) shows the corresponding magnetoconductance MC $= G(H)/G(0)$, where $G(H) = 1/R(H)$ and $R(H)$ is the electrical resistance under magnetic field. Beyond 0.1 T the MC presents an approximately linear dependence with H but a slight MC curvature is also present.

To further characterize the behavior of the magnetic field dependent transport properties, the magnetoconductance was fitted with a second order polynomial of the form [4]: MC $= a(T) + b(T)H + c(T)H^2$. The field-independent parameter $a(T)$ is related to the low-field MC and depends on the magnetization of the samples (M_{film}) [5], while the linear coefficient $b(T)$ is associated with a constant GB susceptibility χ_b [6]. The quadratic term accounts for the curvature at the higher fields [4, 5] and is associated with a GB magnetization (M_{GB}) [1]. In the case where M_{GB} is neg-

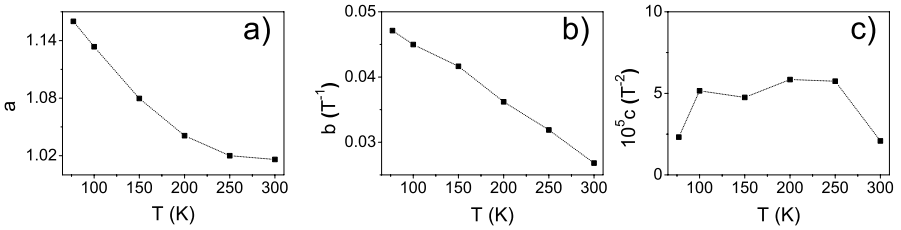

Fig. 3 Parameters obtained from the fitting of a second order polynomial to the measured magnetoconductance

ligible then the magnetoconductance is purely linear and no quadratic term appears on MC.

Figure 3 shows the temperature dependence of the *a*, *b* and *c* parameters obtained from the fittings. As the temperature increases *a* and *b* strongly decrease while *c* is approximately constant on all the measured temperature range. This approximately constant behavior of *c* is indicative of antiferromagnetism on the grain boundary spins [4]. On the other hand, the decrease of *a* and *b* with increasing temperature is due to the decrease of the films magnetization and the increase of inelastic tunneling (with spin-polarization loss) [7], as one progressively approaches T_C.

In conclusion, thin films of $La_{0.67}Sr_{0.33}MnO_3$ were deposited by pulsed laser ablation on $LiNbO_3$ substrates and their electrical properties were characterized up to high magnetic fields. From the fittings to the magnetoconductance, the grain boundary contribution to the magnetic field dependent transport properties was determined. The observed temperature dependence of the fitting parameters was indicative of antiferromagnetism across GB spins and reflected the progressive reduction of the nanograins magnetic ordering with increasing temperature.

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